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OSCILLATION MECHANISM IN CO_2

N. N. Sobolev and V. V. Sokovikov†

ABSTRACT: The processes which produce population inversion of the vibration modes of a CO_2 molecule in CO_2 lasers are considered. On the basis of the experimental works by Schulz, it was assumed that the main process responsible for the upper laser level population is vibrational excitation of N_2 and CO molecules by electron impact and the resulting resonance transmission of oscillation energy to CO_2 molecules. There can be direct 00°1 CO_2 level excitation by electrons as well, although the above-mentioned process alone is able to produce a pumping rate corresponding to the experimentally observed values of specific oscillation power. It is proved that the excitation rate of the lower laser level is described by the relaxation of the bending vibration mode. Hence, the introduction of foreign gases into the discharge tube, which increases the rate of the 01^10 CO_2 level destruction, is expected to produce an increase of the inversion laser level population. Addition of helium to CO_2 leads, above all, to a gas temperature drop, which in turn reduces the rate of the upper laser level destruction.

The continuous emission of coherent radiation in a number of vibration-rotation CO_2 bands in the 10 μm region was discovered in 1964 [1]. The generated power was 1 mW. Similar results were obtained in [2] at almost the same time. In about one year, a power of 10 W had been attained; it was concentrated in two vibration-rotation lines [3]. At a conference on quantum electronics at Phoenix (USA) in 1966, a number of scientists indicated that they had attined powers greater than 100 W. According to the data published, powers which exceed 1 KW, with an efficiency of about 15%, have now been attained [4]. /401*

Such unusually great powers and efficiencies for gas lasers operating in a continuous regime naturally arouse a great amount

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* Numbers in the margin indicate pagination in the foreign text.

of interest in CO_2 lasers from the point of view of applied as well as theoretical physics. The achievements attained by analyzing and studying CO_2 lasers have been described in surveys [5].

In this article, we will discuss only the oscillation mechanism in CO_2. In explaining the function of any gas laser, including a CO_2 laser, two questions must be answered: (1) Which processes guarantee pumping to the upper laser level; (2) Which processes cause destruction of the lower laser level population? The physical aspects of the operation of CO_2 lasers mentioned below were based on data treating descriptions of various processes in theoretical and experimental studies. /402

PROCESSES WHICH PRODUCE THE UPPER LASER LEVEL POPULATION OF CO_2 MOLECULES

Figure 1 gives a schematic diagram of the vibration levels of a CO_2 molecule. The 00°1 level is the upper laser level. The vibration-rotation transitions to the 10°0 and 02°0 levels are laser transitions. It was established in works by Patel [1, 3] that the principal factor causing the inverse population of the upper 00°1 laser level of CO_2 molecules in a CO_2-N_2 laser is the resonance transmission of energy from N_2 molecules in the first vibration level. However, his preliminary explanation of the large population of the vibration levels of the basic N_2 electron state due to electron-ion and atom recombinations, as well as cascades with ex-

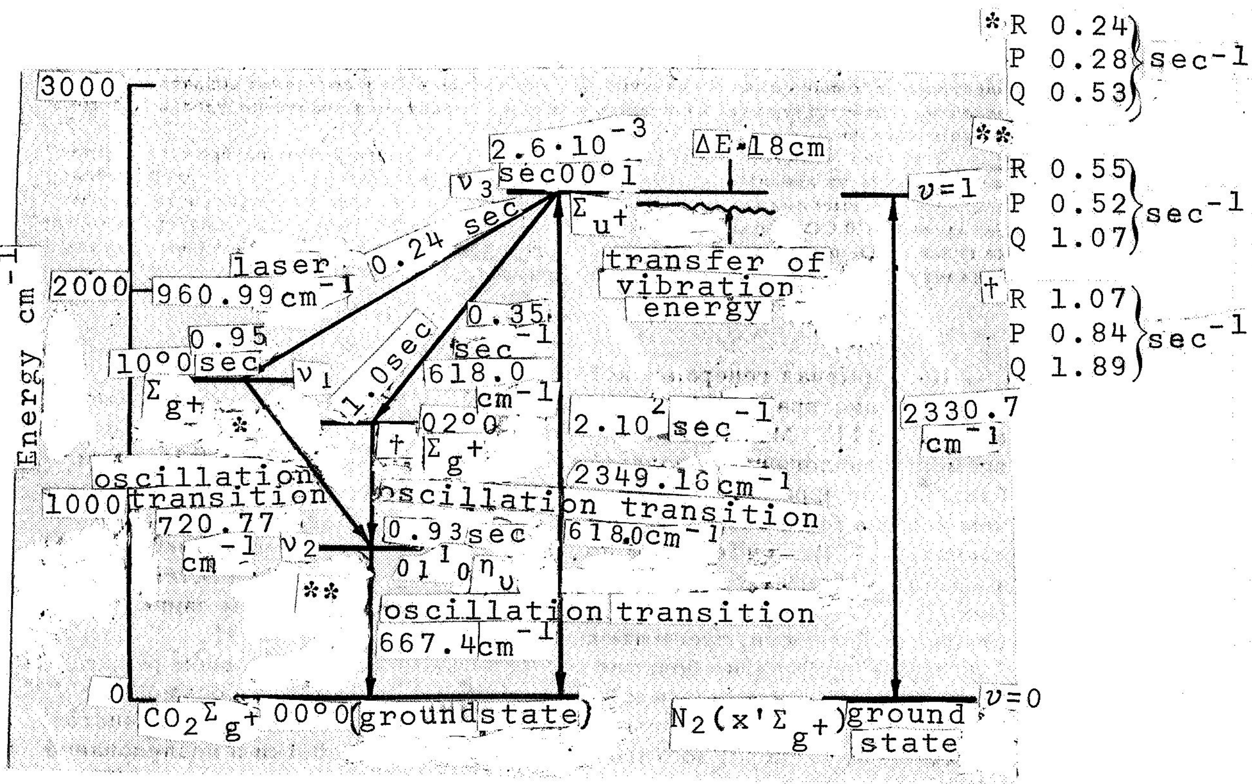

Fig. 1. Schematic Diagram of the Lower Vibrational Levels of CO_2 and N_2 Molecules.

cited electron states, was unsubstantiated and very unlikely. Moreover, Patel's interpretation cannot explain the high power generated in sealed CO_2 and CO_2-He lasers.

In another study [6], we proposed a much more plausible hypothesis in terms of a direct electron excitation of N_2 and CO vibration levels; this is the basic process which causes the large population of the upper laser level in a CO_2 laser. Having made this assumption, we not only could understand the possibility of attaining a high generated power in CO_2-N_2 lasers, but also could explain why the values of the powers and efficiencies attained in sealed CO_2 and CO_2-He lasers are comparable to the powers of CO_2-N_2 lasers. /403

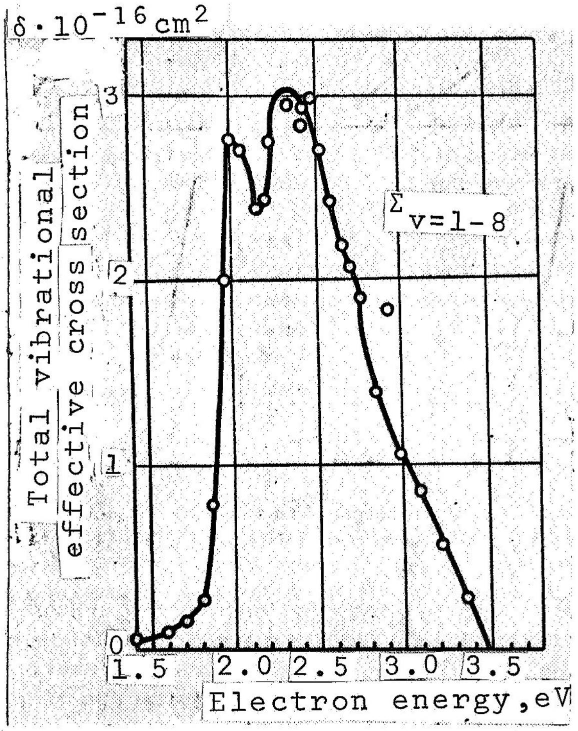

Fig. 2. Dependence of the Cross Section of Vibrational Excitation of N_2 Molecules on the Electron Energy.

Our hypothesis was based on studies by Schulz [7] and Swift [8]. Schulz studied inelastic collisions of electrons with N_2 and CO molecules, which led to vibrational excitation of these molecules. He maintained that the corresponding effective cross sections (see Figures 2 and 3) have a resonance nature and reach a maximum at electron energies of 2.3 eV [$\sigma(e, N_2) = 3 \cdot 10^{-16}$ cm^2] and 1.76 eV [$\sigma(e, CO) = 8 \cdot 10^{-16}$ cm^2]. We can see from Figures 2 and 3 that the absolute values of the total cross sections involving the excitation of the vibrational levels up to the eighth level are very high for electron energies from 1.7 to 3.5 eV in the case of N_2, and for electron energies from 1 to 3 eV in the case of CO. Schulz measured the partial as well as the total cross sections of individual levels. He found that the excitation cross sections of the first to fourth levels are comparable, while the excitation cross section of the seventh and eighth levels is one order less. The interpretation of the high values for the cross sections of the electron excitation of the vibrational levels and their resonance dependence on the energy was linked with the formation of short-lived negative N_2^- and CO^- ions. Theoretical calculations of the cross sections have shown a very satisfactory agreement with Schulz' experimental data.

Figure 4 shows the results Swift obtained by studying the distribution of electrons by energies in a positive glow-discharge N_2 column [8]. We can see that the distribution of electrons by energies is clearly not the Maxwell distribution. It has a maximum /404

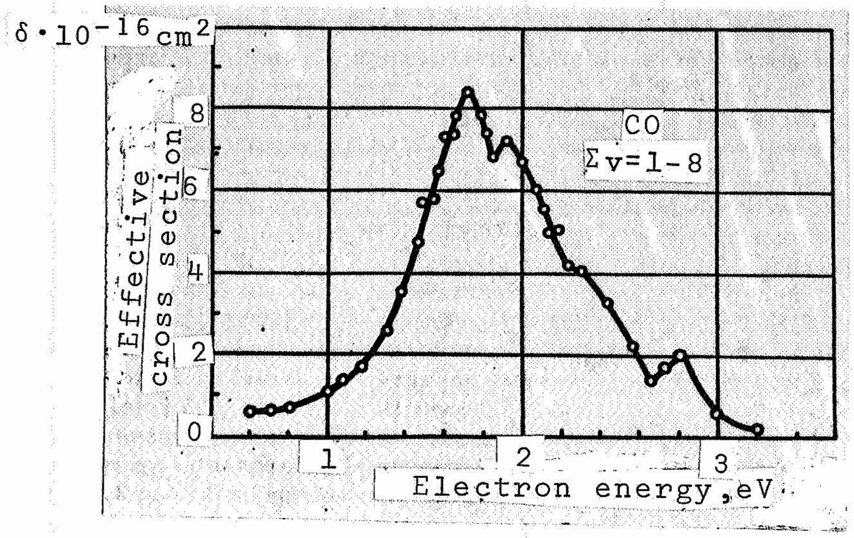

Fig. 3. Dependence of the Cross Section of Vibrational Excitation of CO Molecules on the Electron Energy.

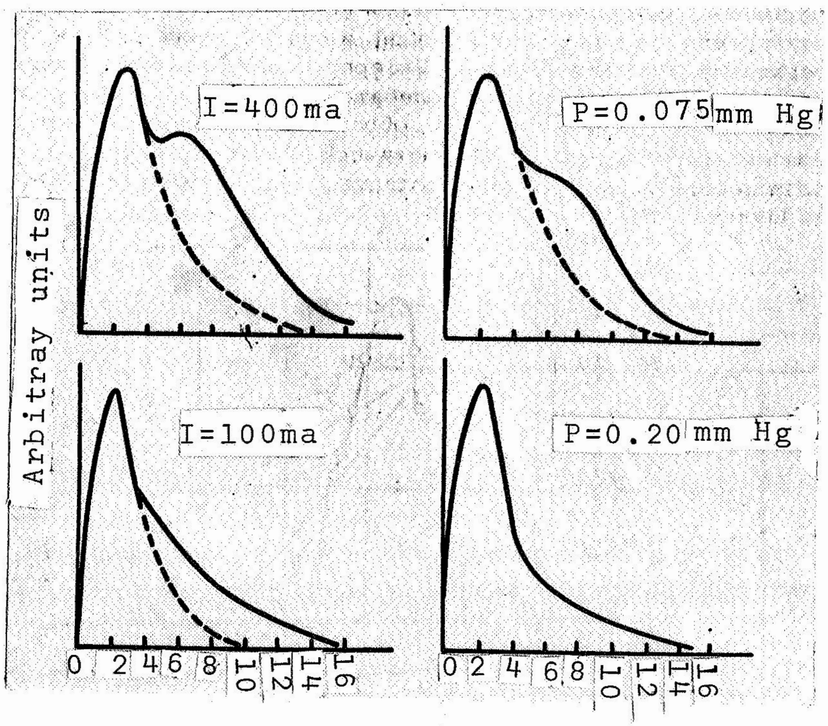

Fig. 4. Functior of the Distruction of Electrons by Energies in a Positive Glow-Discharge N_2 Column.

for an energy of 2-3 eV; with an increase of pressure, there is a shift from the maximum to lesser energies, and the number of fast electrons decreases significantly. This result agrees with Schulz' data. The decrease in the number of electrons with energies close to 1.5 eV was caused by the resonance interaction between an electron and N_2. The decrease in the number of fast electrons with an increase in the number of N_2 molecules (with an increase of the pressure) was caused by an increase in the number of transmissions of energy from the electrons to the N_2. In the case of a CO_2-N_2 laser, in which the operations are conducted at greater pressures than in Swift's experiments, it is safe to say that the average electron energy is no greater than in Swift's experiments. Thus, the average electron energy, under conditions similar to those in CO_2-N_2 lasers, is $\sim$ 2-3 eV (see [9]). Considering the high value of $\sigma(e, N_2)$, this means that the principal cause of the significant N_2 concentration in the excited vibration states is the direct electron excitation of the vibrational levels. /405

We should mention that the effective use of vibrationally excited N_2 for populating the upper CO_2 laser level is possible, not only for values of the vibrational quantum number v = 1, but also all the way to the values of v = 4; the anharmonicity of an N_2 molecule for these values of v does not lead to values of the vibrational quanta which differ from the energy of the 00°1 CO level more than T does. The possibilities of an effective use of a group of vibrational N_2 levels for exciting the upper CO_2 laser level favors the attainment of high values for the efficiency and power of a CO_2-N_2 laser.

It is safe to say that direct electron excitation of a group of vibrational N_2 levels (v = 1-8), and the subsequent resonance transmission of energy by the vibrational CO_2 levels (00°v), can produce values for the rates of the upper CO_2 laser level population (00°1) such that these rates will be sufficient for a high output of generated power.

In order to evaluate this, we will assume that the electron concentration in the glow discharge in a CO_2-N_2 laser is on the order of 10^{10} cm^{-3}. Averaging the effective cross sections of the vibrational excitation of molecules according to the Maxwell distribution of the rates, we will find that, for an electron temperature of 20,000° K and a N_2 pressure of 4 mm Hg, the excitation rate of the vibrational nitrogen levels is $5 \cdot 10^{18}$ sec^{-1}. A consideration of the above-mentioned possibility of excitation of higher vibrational N_2 levels indicates that the excitation rate can increase roughly by a factor of two.

The characteristic value of the specific generated power of a CO_2-N_2 laser is 10^6 erg cm^{-3} sec^{-1}, which corresponds to $\sim 5 \cdot 10^{18}$ excitations per second [10]. Therefore, we can consider that the principal process which produces the upper laser level population is the direct vibrational electron excitation of N_2 molecules, with a subsequent resonance transmission of quanta by CO_2 molecules. Naturally, we have not excluded the possibility of pumping to the upper 00°1 CO_2 laser level by other processes (in particular, direct electron excitation of the upper 00°1 CO_2 laser level). The possibility that this process can be highly effective is seen in the curve for the probability of impacts between electrons and CO_2 molecules at low energies [11]. We could mention the earlier works of Terenin, Nauyman [12] and Dodonova [13] in this respect. However, we cannot draw a single conclusion for N_2 and CO molecules at this time. Each additional process of excitation of the 00°1 level can only improve the conditions for emission in some way, and increase the output power of the CO_2 laser. /406

Earlier, in discussing these aspects, we suggested that there is a strong impact relationship between the N_2 and CO_2 levels. This idea is well known; in the case of CO_2 molecules, it was also confirmed by the work of Hocker et al. [14]. It was found that, after a rapid depletion of the 00°1 vibrational level of a CO_2 molecule, it was again filled within 10^5 sec because of the high population of the overlying layers. The combination of several layers of asymmetrical types of vibrations is also the result of that reservoir, which can support the high population of the upper 00°1 laser level.

In discussing the oscillation mechanism in pure CO_2, we must keep in mind that, as a result of the low energy of CO_2 dissociation (2.8 eV), a significant quantity of CO molecules can be formed in an electron discharge. Considering the large cross sections of excitation for vibrational levels with electron impact, as well as the fact that the difference in the energies of the CO vibrational quantum and the upper CO_2 laser level is within the limits of ($\Delta E_{CO_2,CO} = 170\ cm^{-1}$), we can assume that CO has the same role as N_2 in a CO_2-N_2 mixture. The reduced effectiveness of generation in pure CO_2 (actually, a CO_2-CO mixture) is also understandable, since $\Delta E_{CO_2,CO} > \Delta E_{CO_2,N_2}$.

The hypothesis on the direct electron excitation of N_2 and CO as the principal process for pumping to the upper CO_2 laser level is in good agreement with the high efficiency. The highest theoretically admissible efficiency of a CO_2 laser is equal to the following:

$$\eta = \frac{E_{00^\circ 1} - E_{10^\circ 0}}{E_{00^\circ 1}} \approx 0{,}38,$$

On the other hand, the highest experimental efficiency is about 0.30. There is no doubt that this high value for the efficiency can be attained only by a direct electron pumping which has (in contrast to cascade processes) the least amount of intermediate energy losses.

Thus, the hypothesis based on experimental facts concerning direct electron excitation of the CO and N_2 vibrational levels can aid in explaining the operation of a CO_2-N_2 laser, as well as a CO_2 laser, and their high efficiencies and powers. Therefore, the hypothesis of direct electron excitation of N_2 and CO vibrational levels is also more reliable than assumptions in terms of cascade processes.

PROCESSES RESPONSIBLE FOR DESTRUCTION OF THE VIBRATIONAL LEVELS OF CO_2 MOLECULES

While pumping to the upper CO_2 laser level is assured by electron collisions (the principal process determining the inverse population of the laser levels), there are processes of destruction of the lower level of deformation types of vibrations as a result of intermolecular collisions. This assumption was discussed in a previous article by us [15], as well as in many studies by other authors [16-18].

Figure 5 shows the temperature dependence of the average number of gas-kinetic collisions Z_{10} of CO_2 molecules necessary for the /407

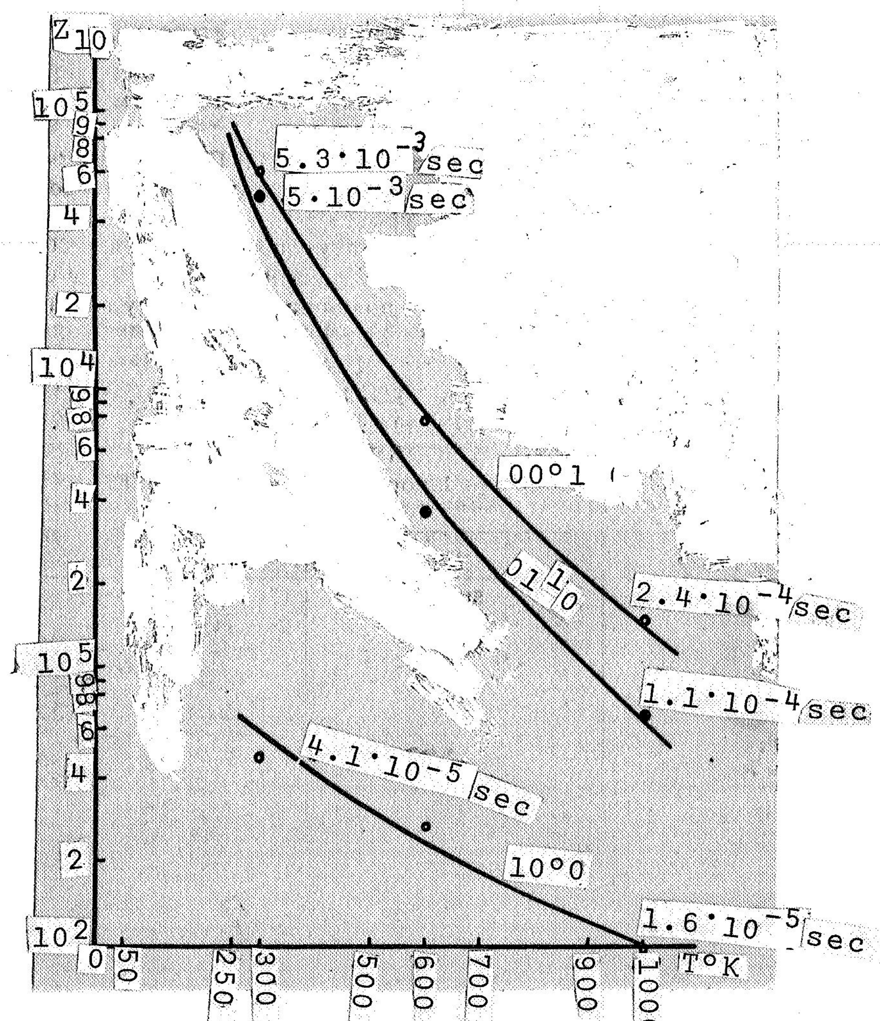

Fig. 5. Temperature Dependence of the Average Number of Gas-Kinetic Collisions Z_{10} Necessary for Destruction of the 10^o0, 00^o1 and 01^10 Levels of CO_2 Molecules.

destruction of the 10^00, 00^01 and 01^10 vibrational levels [19-22]. The corresponding relaxation times τ for a CO_2 pressure of 1 mm Hg are also shown for temperatures of 300° K and 1000° K. As we can see from the figure, the times τ of the 00^01 and 01^10 levels have one order of magnitude. The relaxation time for the 10^00 level (below the laser level) is two or one order(s) less.

If we consider that the characteristic time for transfer between vibrational quanta of one type of vibration is on the order of 1010^{-6}-10^{-5} sec, and that the conversion of vibrational energy (for all types of vibration) into kinetic energy can occur mainly by relaxation of deformation vibrations, we can draw the very important conclusion that high values for the inverse population level and power cannot be attained in pure CO_2. The relaxation time of the 01^10 level will limit both the inverse and the boundary oscillation power. Table 1 gives data on the effectiveness of destruction of the 01^10 level due to collisions with a number of molecules and atoms [20]. We can see that the addition of foreign gases to CO_2 at a partial pressure of 1 mm Hg increases the rate of 01^10 CO_2 destruction by a factor of one to three, so that the preventative role of the 01^10 CO_2 level is eliminated. /408

It becomes clear from this that the CO and N_2 in a CO_2 laser are the molecules which produce the pumping, and are very essential for destruction of the lower laser level.

The significance of adding helium to the gas discharge plasma of a CO_2 laser mainly involves an increase in the rate of the 01^10 CO_2 level destruction (see Table 1). This explains the possibility of obtaining high specific powers.

TABLE 1. AVERAGE NUMBERS OF COLLISIONS Z_{10}, CAUSING A LOSS IN THE VIBRATIONAL QUANTUM, AND THE RELAXATION TIME τ OF THE 01^10 CO_2 LEVEL IN THE PRESENCE OF AN ADDED GAS (PRESSURE, 1 mm Hg; TEMPERATURE, 300° K).

Added gas	Z_{10}	τ, sec
CO_2	50 000	$4{,}4\cdot10^{-3}$
N_2	1200	10^{-4}
CO	230	$2{,}0\cdot10^{-5}$
NO	260	$2{,}2\cdot10^{-5}$
H_2	300	$2{,}6\cdot10^{-5}$
H_2O	60	$5{,}2\cdot10^{-6}$
He	2600	$2{,}2\cdot10^{-4}$

The assumptions we made explain the observed inversions ($\sim 10^{15}$ cm^{-3}) and the specific powers ($\sim$ 0.1 W) obtained in a CO_2 laser.

In order to evaluate them, we will assume that we have the following mixture: 3 mm Hg for CO_2, 3 mm Hg for N_2, and 4 mm Hg for

He. For a pumping rate of $\sim 10^{19}$ cm^{-3} sec^{-1}, we will obtain populations of the upper $00^{0}1$ CO_2 laser level equal to $\sim 2 \cdot 10^{16}$ cm^{-3} at T = 300° K, and $8 \cdot 10^{14}$ cm^{-3} for T = 1000° K. The relaxation time of the $01^{1}0$ level is about $6 \cdot 10^{-5}$ sec, and the rate of population destruction will not limit either the level of the inversions obtained or the level of the specific power. We should mention that helium is significant in other ways than merely accelerating the $01^{1}0$ level destruction. Recent experiments [23] have shown that the addition of He to a CO_2+N_2 mixture decreases the gas temperature T from 800-1000° K to 500-600° K, and to 400° K with water cooling. This means that the destruction of the upper laser level due to collisions with CO_2 molecules decreases, the inversion increases, and the possibility of a more effective use of pumping to the upper level increases. The rate of the $01^{1}0$ level destruction does not decrease, since Z_{10} does not depend on temperature to a great extent in collisions with He atoms.

On the basis of the physical aspects formulated above, we proposed a method for calculating the populations of the vibrational levels of a CO_2 laser [24]. Data for a quantitative comparison between theory and experiment can be obtained with this method.

CROSS SECTIONS NECESSARY FOR AN EXHAUSTIVE EXPLANATION OF THE OPERATION OF A CO_2 LASER

/409

The main physical aspects involved in the operation of a CO_2 laser have been mentioned above. However, for an exhaustive explanation and for reliable calculations, the cross sections of the following processes are necessary:

(1) Dissociation of CO_2, N_2 and CO molecules during an electron impact;

(2) Excitation of all three types of normal vibrations of a CO_2 molecule by an electron impact;

(3) Destruction of the upper CO_2 laser level ($00^{0}1$) due to collisions with CO and N_2 molecules and He atoms at various gas temperatures;

(4) Resonance transmission of the oscillation energy from CO and N_2 molecules to asymmetrical types of vibrations of a CO_2 molecule, and its temperature dependence;

(5) Transfer by quanta of one specific type of CO_2 vibration.

Data on these sections will aid in formulating a complete physical picture of the operation of a device as important as a CO_2 laser.

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